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High Yield Magnetic Nanoparticles Filled Multiwalled Carbon Nanotubes Using Pulsed Laser Deposition.

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Abstract— We present a high yield filling technique of multiwalled carbon nanotubes (MWNTs), grown vertically on a SiO₂ substrate, with magnetic nanoparticles using pulsed laser deposition (PLD). Magnetization measurements in-plane and out-of-plane with respect to the sample surface indicate reasonable coercivity estimated at 0.4 T. The magnetic anisotropy is however found to be randomly oriented, indicating a polycrystalline structure. The unique difference between the in-plane and out-of-plane magnetizations is the sharing produced by the demagnetizing field in the perpendicular direction.

BACKGROUND

Since its discovery in the early 1990s [1], there has been exponential increase in research on carbon nanotubes (CNTs) due to its potential for applications in a wide range of technologies. Of particular interest has been to realize CNTs impregnated with functional materials, such as the magnetic nanoparticles, which are of interest in electronics and data storage. A major technical challenge toward that has been the the assembly of ordered nanoscale structure and controlled filling. In recent years, there has been success in preparing vertically aligned nanotubes on SiO₂ substrate by chemical vapor deposition (CVD)[2]. In this paper, we report a single step procedure to fill vertically aligned multiwalled carbon nanotubes (MWNTs) with cobalt-ferrite using pulsed laser deposition (PLD).

There has been a previous attempt to fill MWNTs in aqueous suspension with cobalt ferrite [3]. Recently, we reported successful chemical filling of CNTs with ferrite nanoparticles [4]. The present work is the first attempt ever to fill MWNTs using PLD, a technique which is commonly used to prepare magnetic thin films. The method presented here offers technologically useful, aligned MWNTs, which are suitable for functional devices.

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EXPERIMENT

Vertically aligned MWNTs were grown by CVD technique on SiO₂ substrate following the procedure of Wei et al [2]. This method involves exposing silica structures to a mixture of ferrocene and xylene at 770 °C for 10 min. The furnace is pumped down to ~200 mtorr in argon bleed and then heated to the temperature of 770 °C. The solution of ferrocene dissolved in xylene (~0.01g/ml) is pre-heated in a bubbler to 175 °C and then passed through the tube furnace. The furnace is then cooled down to room temperature.

The filling material CoFe₂O₄ has a great technological interest because of its large anisotropy and magnetostriction. The filling was carried out in high vacuum (2×10^{-7} Torr) where the polycrystalline CoFe₂O₄ was ablated with a pulsed excimer laser (KrF) at 1.5 J/cm² and 3Hz as energy density and repetition rate respectively. During deposition the SiO₂ template was heated at 300°C and the target was rotated in order to ensure its uniform wear. A total of 12,000 shots were fired to fill the nano-tube structure.

RESULTS

Scanning electron microscope (SEM) image of the vertically aligned MWNTs grown on SiO₂ substrate before deposition is shown in Fig. 1. As shown in the figure, although the majority of the tubes are aligned vertically, a few are misaligned.

In Fig. 2, SEM of vertical tubes after being filled with cobalt ferrite is shown. The filling depth of the magnetic material in the vertically aligned tubes is not apparent from the SEM image. However, it is possible to obtain this information from the in-plane and out-of-plane magnetization measurements on the sample.

Magnetization measurements were performed with vibrating sample magnetometer, both in-plane and out-of-plane with respect to the sample surface. The result of the magnetization measurements are shown in Fig. 3, depicting the magnetic moment as a function of applied field. The hysteresis loop with solid line correspond to the in-plane and that with scatter points correspond to the out-of-plane magnetization. As is clear from Fig. 3 both loops indicate reasonable coercivity estimated at 0.4 T. However, the magnetic anisotropy appears to be randomly oriented (polycrystalline) since both loops exhibit the same hysteresis. The unique difference between the in-plane and out-of-plane magnetizations is the sharing produced by the demagnetizing field in the perpendicular direction.

SUMMARY

In summary, we have, for the first time, used pulsed laser deposition technique for high-yield magnetic nanoparticles filling of vertically aligned carbon nanotubes. The magnetization measurements suggest polycrystallinity of the filled magnetic nanoparticles, as evidenced from the randomly oriented magnetic anisotropy. We believe that our present work further extends the applications of CNT-based materials in electronics technologies.

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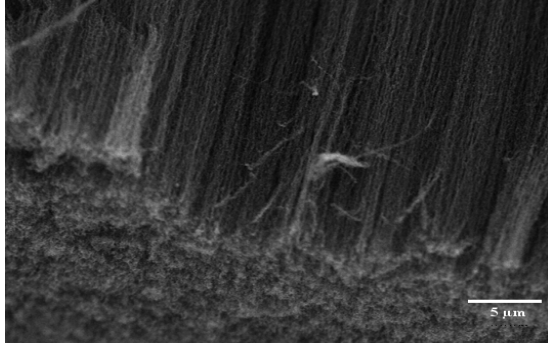


Figure 1: SEM micrograph of vertically grown MWNTs on SiO₂ before filling.

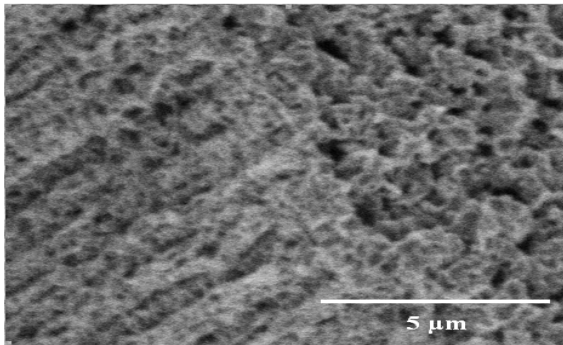


Figure 2 : Top view SEM image of vertically grown MWNTs on SiO₂ filled with CoFe₂O₄ by PLD at high resolution in (a) and in lower resolution (b).

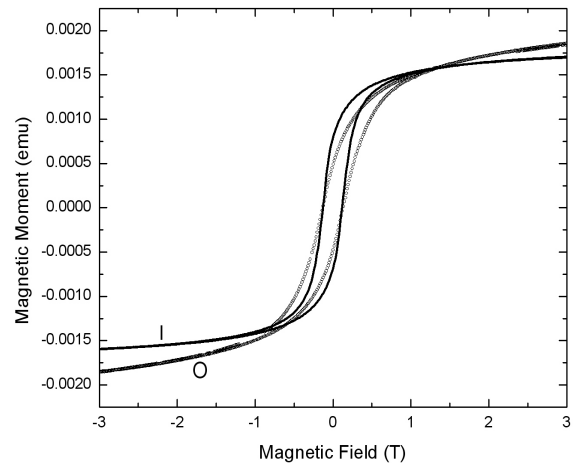


Figure 3: Magnetization of the cobalt ferrite nanoparticles filled, vertically aligned MWNTs. The solid lines represent in-plane and scatter plots represent out-of -plane magnetization.